RADIATION PHYSICS NOTE 112

Method for Estimation of Specific Activities Using Survey Instruments

Vernon Cupps

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INTRODUCTION:

This radiation physics note was undertaken after several attempts to set an empirical basis for characterization of radioactive waste[1] made it clear that a new nomenclature and method was needed. This empirical basis was to be established by correlating readings on survey instruments with a detailed knowledge of typical residual activation products in specific materials.

Over the last two years, various types of activated material were accumulated at the Activation Analysis Laboratory (AAL) and then analyzed on a gamma ray spectrometer to determine the specific activities of each radionuclide contained in the activated material[1]. Before the analyses, contact readings with a Bicron Analyst™ (colloquially known as the Bicron) and a Ludlum 177-4 Frisker™ (colloquially known as the Frisker) were recorded for each item. One must now formulate a method for correlating the gross gamma and beta activities seen with the survey instruments to the radionuclide specific data obtained with the gamma ray spectrometer. That is the purpose of this note.

Theory:

1. General

Radionuclides which decay through multiple modes can radiate several different energy gamma and/or beta rays. These occur either simultaneously or with a frequency defined by the proportion of decays (branching ratio) which give rise to a particular beta or gamma-ray decay mode. Since the specific activity of a material is directly proportional to the number of disintegrations of that material per unit time, each gamma ray will provide an independent determination of that specific activity. The average of the specific activities determined for each gamma ray (defined by its energy) will then yield a mean value for the specific radionuclide activity. This average specific activity is what is needed to characterize waste for shipment. On the other hand, in order to correlate survey meter readings with specific activity, the relative contribution of the individual emissions from a specific radionuclide to the count rate of the survey meter is needed.

Survey instruments are non-specific detectors of radiation. That is, they respond to any gamma or beta ray possessing sufficient energy to register a pulse within the electronic detection window. The energy response to gamma radiation for the Bicron Analyst™ which uses a NaI(Tl) detector is similar to that for the gamma ray spectrometers at the AAL which are LN_2 cooled Germanium detectors. On the other hand, another survey instrument, the Frisker, has a response to gamma ray radiation that is significantly different from that of the Germanium detectors used at the AAL or the NaI(Tl) detectors used in the Bicrons. A primary difference is the intrinsic efficiency with which each type of detector converts gamma rays incident on them into electrical pulses. For energies greater than 120 keV, the response of the Frisker is approximately constant with energy and two orders of magnitude below the efficiency of either

the Bicron's NaI(Tl) detector or the Germanium solid-state detectors. In both the Germanium solid-state and the NaI(Tl) scintillation detectors, the response falls roughly exponentially between 150 keV and 2000 keV. This analogous behavior begins to deviate from ideal near 120 keV where absorption in the dead layer of the Germanium solid-state detectors force the gamma ray detection efficiencies to drop off precipitously while the efficiency for a NaI(Tl) detector remains essentially constant. Fortunately most accelerator produced radionuclides emit gamma rays with energies well above this region of difficulty. One can define the following parameters:

$\varepsilon_F(E)$ = Intrinsic total detection efficiency of Frisker for a gamma ray of energy E.

$\beta_F(E)$ = Beta detection efficiency of Frisker for a beta ray of energy E.

$\varepsilon_G(E)$ = Intrinsic total detection efficiency of HpGe for a gamma ray of energy E.

$\varepsilon_g(E)$ = Intrinsic full peak efficiency of HpGe detector at energy E. This is the efficiency used to calculate the specific activities.

$\varepsilon_B(E)$ = Intrinsic total detection efficiency of Bicron Analyst for a gamma ray of energy E.

$\beta_B(E)$ = Beta detection efficiency of Bicron Analyst at energy E. This is negligible when compared to the gamma detection efficiency.

SA_j = Specific Activity of radionuclide j (pCi/gr) at time t. Note that the time dependence is implicitly understood.

m = Mass of material being analyzed (gr).

B_{ji} = Branching Ratio for i^{th} beta from radionuclide j.

G_{ji} = Branching Ratio for i^{th} gamma ray from radionuclide j.

Ω_g = Average fractional (of 4π) solid angle subtended by HpGe detector at material being counted.

Ω_b = Average fractional (of 4π) solid angle subtended by Bicron Analyst™ detector at material being counted.

Ω_f = Average fractional (of 4π) solid angle subtended by Frisker detector at material being counted.

κ = Conversion constant; 0.037 Bq/pCi.

With these definitions one can now attempt to lay a foundation for using survey instruments, coupled with detailed process knowledge, to establish radionuclide specific information on various materials containing accelerator produced radionuclides.

2. Bicron Survey Meter

Each survey instrument responds to a range of gamma ray energies determined by electronic settings and detector response characteristics. The low energy cutoff for the Fermilab Bicron

survey instruments occurs at approximately 60 keV. Virtually all gamma radiation from long lived accelerator produced radionuclides is less than 2800 keV. Therefore for measurement purposes, ε_G (and ε_B) can be defined by the following equation:

$$\varepsilon_G \text{ (or } \varepsilon_B) \equiv \frac{\text{\# of Counts between 60 \& 2800 keV}}{\text{\# of } \gamma\text{s of energy E incident on Detector}} \qquad \text{(A)}$$

The count rate (CR_B) seen on a Bicron will be:

$$(CR_B)_j = \left\{ \sum_{i=1}^{n} \left(\kappa \cdot \Omega_b \cdot m \cdot SA_j \right) \cdot G_{ji} \cdot (\varepsilon_B)_{ji} \right\} \cdot 60 \qquad \text{(B)}$$

where n is the number of different nuclear transformations which lead to the emission of a gamma ray by radionuclide j. A factor of 60 is necessary to convert from cps to cpm. Note that a correction factor for loss of gamma flux due to self absorption $(F_b)_{ji}$ in the material being counted has not been included in equation (A) since for these measurements it is assumed to be negligible. Note also that the $(CR_B)_j$ are each implicitly time dependent due to the SA_j. If the material being counted contains only one radionuclide then this equation can be solved in a straightforward manner for the specific activity of that radionuclide. However in most materials more than one radionuclide is created through the activation process. These radionuclides are created at different rates and decay at different rates. Hence their relative contribution to the count rate seen on a Bicron is in general a function of time. For the case of multiple radionuclides in a material, the total count rate above ambient background (T_B) can be expressed as a sum of contributions from each of the individual radionuclides.

$$T_B = \sum_{j=1}^{k} (CR_B)_j \qquad \text{(C)}$$

where k is the number of distinct radionuclides detected in the activated material. The relative contribution (R_B) of each radionuclide to the total observed gamma activity seen by the Bicron is then:

$$R_B(t)_j = \frac{(CR_B)_j}{T_B} \qquad \text{(D)}$$

Under the same counting conditions, the constants contained in equation (B), i.e., Ω_b, m, κ, and 60, will be the same for all radionuclides in a given activated material when a homogenous distribution is assumed and will therefore cancel in the expression for the relative contribution R_B. Equation (D) then simplifies to:

$$R_B(t)_j = \frac{\left\{ \sum_{i=1}^{n} (SA_j) \cdot G_{ji} \cdot (\varepsilon_B)_{ji} \right\}}{\sum_{j=1}^{k} \left\{ \sum_{i=1}^{n} (SA_j) \cdot G_{ji} \cdot (\varepsilon_B)_{ji} \right\}} \qquad \text{(E)}$$

Determination of an analogous ratio from measurements with the HpGe detector will necessitate the introduction of a new efficiency, ε_G , which will be defined as the efficiency with which the HpGe detector sees a pulse between 60 and 2800 keV from an incident gamma ray of less than 2800 keV. This efficiency is different from the intrinsic full peak efficiency, ε_g , used to determine the specific activities and thus must also be empirically determined. The total count rate seen in the HpGe detector due to the j^{th} radionuclide can be generally expressed as:

$$(CR_G)_j = \left\{ \sum_{i=1}^{n} \left(\kappa \cdot \Omega_g \cdot m \cdot SA_j \right) \cdot G_{ji} \cdot (\varepsilon_G)_{ji} \right\} \cdot 60 \qquad (F)$$

For the case of multiple radionuclides in a material, the total count rate above ambient background between 60 and 2800 keV seen on the HpGe detector would be:

$$T_G = \sum_{j=1}^{k} (CR_G)_j \qquad (G)$$

Implicit assumptions included in this expression are that most of the gamma rays incident on the detector from a specific transition deposit at least 60 keV of energy in the HpGe crystal and that contributions from very low concentration radionuclides, i.e., those that constitute less than 1% of the total activity, can be neglected. The relative contribution of each radionuclide to the total observed gamma activity as seen by the HpGe detector can then be defined by the ratio:

$$R_G(t)_j = \frac{(CR_G)_j}{T_G} \qquad (H)$$

Substitution of previously defined values into equation (H) leads to an equation for the HpGe detector similar to equation (E) for the Bicron Analyst™.

$$R_G(t)_j = \frac{\left\{ \sum_{i=1}^{n} SA_j \cdot G_{ji} \cdot (\varepsilon_G)_{ji} \right\}}{\sum_{j=1}^{k} \left\{ \sum_{i=1}^{n} SA \cdot G_{ji} \cdot (\varepsilon_G)_{ji} \right\}} \qquad (I)$$

For a given material this expression will generate k linear equations for k unknown specific activities at some storage time (t), i.e., time elapsed from the removal of the material from an area where activation occurs, when both the total activity, T_G, and the activity ratio, $R_G(t)_j$, are known. It is these numbers, T_G, R_G, and ε_G,, along with the specific activity, which we need to determine in ref. (1) for a number of storage times. As a check, the total detection efficiencies can be used to calculate a predicted T_G which can then be compared to the measured T_G.

Now the crucial question is, how does one correlate R_B with R_G? If the Bicron Analyst™ total detection efficiency is proportional to the total detection efficiency of the HpGe detector, it can be expressed as:

$$\varepsilon_B = c \cdot \varepsilon_G \qquad (J)$$

This equality depends strongly on the caveats that the electronic detection windows are the same for the gamma ray spectrometer and the Bicron Analyst™, and that both instruments exhibit the same qualitative response to gamma radiation, i.e., that the different functional form of the efficiencies for the two instruments between 60-120 keV is negligible when compared to the full 60-2800 keV range. If the second caveat does not hold then the relationship expressed by equation (J) becomes a more complicated function of energy. Substitution of equation (J) into equation (E), allows the relative observed gamma activity for a Bicron Analyst™ to be approximately expressed as a function of the HpGe detector total detection efficiency:

$$R_B(t)_j = \frac{\left\{\sum_{i=1}^{n} (SA_j) \cdot G_{ji} \cdot (\varepsilon_G)_{ji}\right\}}{\sum_{j=1}^{k}\left\{\sum_{i=1}^{n} (SA_j) \cdot G_{ji} \cdot (\varepsilon_G)_{ji}\right\}} \qquad (K)$$

The validity of equation (J) must be experimentally checked before equation (K) can be used. If it and the accompanying assumptions are correct then

$$R_B(t)_j = R_G(t)_j \qquad (L)$$

and

$$T_B \propto T_G \qquad (M)$$

From a practical point of view all of this hangs together very well if one can find enough pure monoenergetic gamma ray sources to establish a functional relation for ε_G and ε_B. Unfortunately most of the commonly available sources such as ^{60}Co and ^{22}Na emit several different energy gamma rays. Sorting out contributions to the Compton continuum from each of the gamma rays separately is a formidable task. Therefore, in general, ε_G and ε_B are not easily determined. This problem can be bypassed by combining G_{ji} and $(\varepsilon_{G\text{ or }B})_{ji}$ into a radionuclide specific efficiency, $(\varepsilon^R_{G\text{ or }B})$, for detecting a given radionuclide, i.e.,

$$\sum_{i=1}^{n} G_{ji} \cdot (\varepsilon_G \text{ or } \varepsilon_B)_{ji} \cong (\varepsilon^R_G \text{ or } \varepsilon^R_B)_j \qquad (N)$$

Thus the radionuclide specific detection efficiency can now be defined as:

$$\varepsilon^R_G \text{ (or } \varepsilon^R_B) \equiv \frac{\text{\# of Counts between 60 \& 2800 keV}}{\text{\# of } \gamma\text{s from radionuclide R incident on Detector}} \qquad (O)$$

This composite efficiency can then be substituted into equations (E), (I), and (K) to determine the ratios R_B and R_G. For example equation (K) would simplify to:

$$R_B(t)_j = \frac{SA_j \cdot (\varepsilon^R_G)_j}{\sum_{j=1}^{k}\{SA_j \cdot (\varepsilon^R_G)_j\}} \qquad (P)$$

3. Frisker Survey Meter

It is possible that this method can be extended to the gas filled detectors at some time in the future, therefore for the sake of completeness a discussion outlining application of this method to the Frisker survey instrument is included. A much different response to radiation is observed with the Frisker. It responds to both gamma and beta radiation but because it is a thin window GM detector, it primarily responds to the beta radiation from a material during contact surveys. The Frisker contact counting rate in cpm due to gamma and beta flux from radionuclide j or the radionuclide specific observed total activity $\{(CR_F)_j\}$ can be approximately given by:

$$(CR_F)_j = \left\{\sum_{i=1}^{n} (\kappa \cdot \Omega_f \cdot m \cdot SA_j) \cdot G_{ji} \cdot (\varepsilon_F)_{ji} + \sum_{i=1}^{p} (\kappa \cdot \Omega_f \cdot m \cdot SA_j) \cdot B_{ji} \cdot (\beta_F)_{ji}\right\} \cdot 60 \qquad (Q)$$

where n is the number of gamma rays emitted by radionuclide j, p is the number of beta rays emitted by radionuclide j. It should be noted that no attempt has been made to account for loss of gamma or beta flux due to absorption effects. For small volume materials this effect should be negligible but it will introduce large reductions in the predicted survey instrument count rate for items like 55 gal. drums packed with materials which emit beta or low energy gamma radiation copiously. Unlike the Bicron, the Frisker responds more strongly to beta radiation than to gamma radiation. Typically, the efficiency of the Frisker for detecting beta radiation is two orders of magnitude higher than its efficiency for detecting gamma rays. Hence,

$$\beta_F \gg \varepsilon_F \tag{R}$$

and when B_{ji} and G_{ji} are comparable, the net observed Frisker count rate reduces to:

$$(CR_F)_j = \left\{ \sum_{i=1}^{p} \left(\kappa \cdot \Omega_f \cdot m \cdot SA_j \right) \cdot B_{ji} \cdot (\beta_F)_{ji} \right\} \cdot 60 \tag{S}$$

The total observed count on the Frisker would be:

$$T_F = \sum_{j=1}^{k} (CR_F)_j \tag{T}$$

where k is the number of distinct radionuclides detected in the activated material. The relative contribution (R_F) of each radionuclide to the total observed beta activity seen by the Frisker is then:

$$R_F(t)_j = \frac{(CR_F)_j}{T_F} \tag{U}$$

The constants contained in equation (S), i.e., Ω_f, m, κ, and 60, are again approximately the same for all radionuclides in a given activated material and therefore cancel in the expression for the relative contribution R_F. Equation (Q) then simplifies to:

$$R_F(t)_j = \frac{\left\{ \sum_{i=1}^{p} (SA_j) \cdot B_{ji} \cdot (\beta_F)_{ji} \right\}}{\sum_{j=1}^{k} \left\{ \sum_{i=1}^{p} (SA_j) \cdot B_{ji} \cdot (\beta_F)_{ji} \right\}} \tag{V}$$

For a given material this expression will generate k linear equations for k unknown specific activities at some storage time (t) before the measurement. A key difference between this equation and equation (E) is that the total detection efficiency of the Frisker as a function of the beta ray energy will not, in general, be qualitatively similar to the total detection efficiency of the HpGe detector for gamma rays so that a simple expression like equation (J) cannot be written.

CALCULATION OF R_G:

Gamma ray spectroscopy is one method which can be used to empirically determine the relative contribution of each radionuclide to a given material's gamma radioactivity[1]. This procedure can be summarized by the following steps:

- Establish an average natural background (B_g). When possible this should be done using a virgin piece of the same material.

•The activated material should be counted for the same amount of time as the background.

•The background should then be subtracted peak by peak from the spectra generated by the analyzed material.

•Peaks for each distinct gamma ray transition should then be identified and the sums extracted.

•Specific Activities for each radionuclide represented in the spectrum should be calculated from each respective transition. If a given radionuclide exhibits more than one gamma transition, then the specific activities determined from each transition should be averaged together to determine the measured specific activity for that specific radionuclide.

To this point, a typical gamma ray analysis has been described. To calculate R_G consider the following steps:

•The total observed gamma activity above ambient background, T_G, should be empirically determined by summing the entire background subtracted spectrum from 60 keV to 2800 keV. This parameter, along with the specific activities, must be determined for each type of material at a number of different storage times . For example, consider a piece of uniformly activated Al. After the ambient background is subtracted from the Al spectrum, a full spectrum sum from 60 keV to 2800 keV would be recorded. This is T_G.

•Radionuclide specific detection efficiencies, ε_G^R, must be empirically determined using NIST traceable single radionuclide sources. This is the only way to distinguish which portions of the Compton background are due to a particular radionuclide. For the particular example chosen above, a ^{22}Na source would be a suitable choice. The radionuclide specific detection efficiency being determined is defined in equation (O).

•Once the total detection efficiencies of the HpGe detector for various accelerator produced radionuclides have been established, a second method of determining T_G is available. When the denominator of equation (I) is multiplied by the constant term $\{60 \cdot \kappa \cdot \Omega_g \cdot m\}$, T_G should be the result. If these T_G's are not reasonably close to those determined as discussed directly above, then significant contributions to the spectrum have probably been overlooked. For our chosen example, one would multiply the specific activity of ^{22}Na calculated from standard gamma ray spectroscopy by the radionuclide specific detection efficiency for ^{22}Na, ε_G^R. Since this is essentially the only radionuclide present, the sum over k in the denominator of equation (I) drops out and multiplication by the constant term $\{60 \cdot \kappa \cdot \Omega_g \cdot m\}$ should yield T_G if other significant contributors to the observed gamma flux have not been overlooked.

•$R_G(t)_j$ may now be calculated in a straightforward manner from equation (I) for each radionuclide at storage time t. For our above example $R_G(t)_j = 1$.

CALCULATION OF R_B:

•Take five independent ambient background readings with a Bicron Analyst™ using the meter readout with the Bicron set to slow response. Calculate an average background and a standard deviation from the five measurements.

•Perform a contact survey with the Bicron Analyst™ over the entire surface area of the material being analyzed. Record the highest and lowest readings. For small items these should be approximately the same.

•Subtract the average background from the highest contact reading. This provides an empirical determination of T_B.

•The radionuclide specific detection efficiency, ε_B^R, of the Bicron Analyst™ for specific radionuclides should have been previously established through independent measurements in a fashion similar to that used for the HpGe detector with single radionuclide NIST traceable sources.

•Using these total detection efficiencies, the specific activities for each radionuclide determined from gamma ray spectroscopy on the same material sample, and the constant term $\{60 \cdot \kappa \cdot \Omega_g \cdot m\}$, T_B can be calculated from equation (C). This calculated T_B should be approximately equal to the empirically determined T_B and thus serves as a consistency check between measurement and prediction.

•$R_B(t)$ may now be calculated in a straightforward manner from equations (E) or (P) for each radionuclide at storage time t.

•If the assumption expressed in equation (J) holds true then $R_B(t)$ and $R_G(t)$ should be approximately equal.

APPLICATION:

Field application of this technique involves the direct measurement of only one of the three parameters necessary to determine specific activities from equation (E), i.e., the total count rate above ambient background, T_B or T_F. This means that both the relative contribution of each radionuclide to the observed activity, $R_B(t)$ and $R_F(t)$, and the radionuclide specific detection efficiencies for each radionuclide, ε_B^R and β_F^R, must be predetermined. If equation (J) is valid then only $R_G(t)$ and ε_B^R or ε_G^R and the proportionality constant c, need to be predetermined for Bicron Analyst™ surveys. It should be noted again that an expression such as equation (J) does not exist for the Frisker survey instrument, so that the determination of $R_F(t)$ is not a straightforward application when using this instrument.

An experimental study which establishes $R_G(t)$ [$R_B(t)$ or $R_F(t)$] for each radionuclide in a specific type of material as a function of time and the radionuclide specific detection efficiencies ε_G^R (ε_B^R or β_F^R) for a given radionuclide must clearly be an integral part of any field application of this technique. This then will be the intended objective of the study begun by ref. (1).

REFERENCES

(1) K. Vaziri, V. Cupps, and A. Elwyn, Isotopic Analysis of Accelerator Induced Radioactivity in Material, Fermilab Radiation Physics Note #98, Interim Report, 9/92.

(2) Glenn F. Knoll, Radiation Detection and Measurement, Second Edition, John Wiley & Sons, New York, NY, 1989.

(3) Nicholas Tsoulfanidis, Measurement and Detection of Radiation, McGraw-Hill Book Company, New York, NY, 1983.

(4) The Health Physics and Radiological Health Handbook, Revised Edition, Edited by Bernard Shleien, Scinta Inc., Silver Spring, MD, 1992.